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Bioelectrocatalysis and surface analysis of gold coated with nickel oxide/hydroxide and glucose oxidase towards detection of glucose

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ABSTRACT

The fabricating of metal oxide thin films onto conducting substrates continues to grow and their potential applications as surfaces for biosensor applications is of significant importance. The correct orientation of glucose oxidase redox enzymes yields very important biosensors capable of selectively detecting D-glucose as a measure of blood sugar for healthy and diabetic risk patients. The electrodeposition of redox enzymes, such as glucose oxidase enzymes, onto gold electrodes chemically pre-modified with nickel oxide was investigated in this work. The surface characterization confirmed the chemical nature, morphology and thin film composition of the modification of bare and modified gold electrodes. The electrodeposition of GOx enzyme onto nickel oxide/hydroxide thin film resulted in a surface with excellent bioelectrocatalytic properties towards the detection of D-glucose. The nickel within the nickel oxide/hydroxide thin film had a Ni(II) oxidation state. A well-defined redox peak of GOx enzyme co-factor ($FAD/FADH_2$) was observed confirming the oriented immobilization onto NiO/Ni(OH)$_2$ conducting surfaces. The amount of GOx enzyme deposited was determined by integrating the charge (Q = 0.368 μC) under the reduction peak and the surface coverage was found to be 1.43×10^{-10} mol. cm^{-2}. A linear plot of electrocatalytic reduction currents against D-glucose concentrations was obtained up to 30.0 mM with a linear correlation coefficient (R^2) of 0.99. The limit of detection (LoD) using $S/N = 3$ was calculated to be 1.54 ± 0.03 mM with a sensitivity of the biosensors was 1.95 ± 0.13 μA.mM.cm^{-2}. The selectivity towards only D-glucose and not ascorbic acid and uric acid was evaluated and the Au-NiO/Ni(OH)$_2$-GOx could not detect 1.0 mM of ascorbic acid and uric acid.

1. Introduction

The interest in bioelectrochemical sensors has increased over the years due to their advantages, such as sensitivity and selectivity, fast response, easy operation and real-time on-line detection. Biosensors have been of interest in various fields, particularly in biochemistry, environmental monitoring and clinical diagnosis [1]. Glucose-based biosensors are the most widely studied and commercially available systems. They have been used for the detection of glucose in clinical, biological, chemical samples and food processing and fermentation [1,2]. This highlights the variety of media where glucose can be detected and hence the continued demand for glucose biosensors. Repetitive analysis in a variety of physiological fluids is one of the most vital and frequent processes in a clinical laboratories [3–8]. Glucose is an important analyte or biomolecule in the human metabolism processes [8,9]. Patients with diabetes are required to monitor blood glucose levels daily. This has led to a growing demand of glucose monitoring biosensors. Therefore, glucose biosensors that are selective, reliable, cheap and easy to operate for continuous monitoring of glucose levels are of importance [5–7].

Biosensors require modification of electrode surfaces with enzymes impart some selectivity and specificity towards the analyte of interest [1,2,9–12]. The challenge is that redox enzymes, such as glucose oxidase, peroxidase, and laccase do not readily exchange electrons with the solid electrode surfaces [13–20]. This is because the redox cofactor such as FAD (flavin adenine dinucleotide), iron porphyrin (Heme), NAD (nicotinamide adenine dinucleotide) are embedded within the large 3D glycoprotein with complex structures. The redox enzyme 3D glycoprotein hinders optimum interaction and electron transfer with the electrode surface [10]. Modification of electrode surfaces is the method used for improving biomolecular interaction with solid electrode surface. Different types materials such as carbon nanotubes, metal oxides

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